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(54) Title: COPOLYESTER ADHESIVE BLEND

(57) Abstract

Copolyester adhesive blends for use particularly as fabric blends. The blends comprise a crystallizable copolyester and a substantially amorphous polyester.

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COPOLYESTER ADHESIVE BLEND

This invention relates to certain copoly-  
ester blends which have been found to be useful as  
adhesives, especially for bonding fabric, fibers and  
5 the like.

Fabric adhesives are often used on fusible  
interlinings, which are materials such as fabric  
which have been coated on one side with a dis-  
continuous pattern of adhesive. When the interlining  
10 is bonded to a face fabric in a garment, it provides  
body and shape to the garment without impairing the  
ability of the fabric to breathe. Fusible inter-  
linings are used in the manufacture of suits, in  
shirt collars and cuffs, and in the waistbands of  
15 trousers. In the manufacture of suits, polycarbonate  
basting threads are frequently used to temporarily  
hold the parts of the suit in place. After the suit  
is completed, a solvent such as perchloroethylene or  
trichloroethylene is used to embrittle the poly-  
20 carbonate thread so that it may be brushed from the  
fabric. Binder fibers are used with nonwovens, which  
are formed from matrix fibers into a web, sheet, or  
mat. The binder fibers give strength to the wet,  
mat, or sheet by bonding it together.

25 Certain polyesters are known to be useful  
for fusible interlining adhesives and binder fibers.  
However, these polymers also tend to have certain  
disadvantages. For example, one polyester of inter-  
est is the copolyester of terephthalic acid, adipic  
30 acid, ethylene glycol and 1,4-butanediol as described  
in U.S. Patent No. 3,699,921. Such polyesters tend  
to block in pellet form and fumed silica must be  
added in significant amounts to make it possible to  
grind this polymer into powder. Excessive amounts of  
35 fumed silica in the powder, however, prevent good  
coatability and good fusion of the powders on the  
fusible interlining fabric when applied with powder

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point applicators. Thus, these adhesives are not easily applied.

5 Amorphous polyesters cannot be used as fusion adhesives in which resistance to dry-cleaning agents and high set-up speed are required. In like manner, those polyesters are undesirable which have too little crystallinity, because they solidify too slowly and consequently do not lose their surface stickiness for long periods of time.

10 Certain low melting, crystallizable polyesters are useful for bonding fabrics at temperatures (120-150°C.) which do not damage the fabrics. These bonds generally show excellent resistance to laundering and dry cleaning treatments. Thus, these 15 materials are useful in film, fusible interlining, or melt blown web form to laminate fabrics in the construction of wearing apparel. However, in applications where high temperature resistance is required, the low melting polyesters are not completely satisfactory.

20 In U.S. Patent 4,131,714 there is described an adhesive composition which is a blend of a crystalline polyester having a very high melting point, e.g., 243°C, and an amorphous polyester. The blend 25 is used as a solution in an organic solvent usually cresylic acid. This alone makes the composition not useful as a fabric adhesive which must be a solid, e.g., a powder. Further, the cresylic acid is probably necessary in order to provide a plasticizer for 30 the very high melting crystalline polyester used so as to make the composition usable at reasonable temperatures. Cresylic acid would be unsuitable in a fabric adhesive because it would attack the fabric and could irritate the skin of a wearer of the fabric. 35 The compositions of this patent are used in electrical equipment.

Thus, the problem to be solved is to provide a fabric adhesive which is easily applied to fabrics and which shows good high temperature resistance.

This problem is solved by providing a

5 particulate blend of

- a) a crystallizable copolyester having a melting point of 80-130°C and a glass transition temperature between 0°C and 20°C and
- 10 b) a substantially amorphous copolyester having a glass transition temperature of 50°C or higher.

The blends contain 10-99%, preferably 95%, of the crystallizable copolyester and 90-1%, preferably 70 to 5% of the substantially amorphous copolyester.

15 Useful crystalline polyesters are derived from an acid selected from the group consisting of terephthalic acid and 1,4-cyclohexanedicarboxylic acid and a glycol selected from the group consisting of 1,4-butanediol, 1,6-hexanediol and mixtures of

20 1,4-butanediol and 1,6-hexanediol, the copolyester also being derived from about 20-60 mole percent of a co-acid or a co-glycol, or about 20-75 mole percent of a combination of a co-acid and a co-glycol, the co-acid and co-glycol being selected from the group

25 consisting of isophthalic acid, C<sub>3</sub>-C<sub>12</sub> aliphatic dibasic acids, aliphatic and cycloaliphatic glycols having 2-8 carbon atoms and diethylene glycol. These polyesters preferably have a heat fusion of 1-10 calories per gram and I.V. (inherent viscosity) of 0.4-1.2, preferably between 0.75 and 0.95.

Crystallizable copolymers of particular interest include 1) poly(butylene terephthalate) copolymers containing 20 mole percent glutaric acid and 45 mole percent diethylene glycol, 2)

35 poly(butylene terephthalate) copolymers containing 30 mole percent glutaric acid and 45 mole percent

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diethylene glycol, 3) poly(butylene terephthalate) copolymers containing 25 mole percent glutaric acid and 40 mole percent diethylene glycol, 4) poly(hexamethylene terephthalate) copolymers containing 20 mole percent isophthalic acid and 20 mole percent 1,4-butanediol, 5) poly(hexamethylene terephthalate) copolymers containing 10 mole percent isophthalic acid and 40 mole percent 1,4-butanediol, 6) poly(hexamethylene terephthalate) copolymers containing 20 mole percent 1,4-butanediol, 7) poly(butylene terephthalate) copolymers containing 50 mole percent isophthalic acid, 8) poly(butylene 1,4-cyclohexanedicarboxylate) copolymers containing 25 mole percent glutaric acid and 25 mole percent diethylene glycol, 9) poly(hexamethylene terephthalate) copolymers containing 20 mole percent glutaric acid and 20 mole percent diethylene glycol and the like.

Useful amorphous copolymers are derived from at least one acid selected from the group consisting of terephthalic acid, isophthalic acid, and 1,4-cyclohexanedicarboxylic acid, and at least one glycol selected from the group consisting of ethylene glycol, diethylene glycol, and 1,4-cyclohexanedimethanol, with the proviso that the copolymer contains at least two acids or at least two glycols, the second acid or glycol being present in an amount of 25-60 mole percent.

Substantially amorphous copolymers of particular interest include polyesters having  $T_g$  values above 50°C, preferably 50-90°C, heats of fusion  $\Delta H_f$  of less than one calorie per gram and inherent viscosities of 0.4 to 1.2. Some typical polymers include 1) poly(ethylene terephthalate) copolymers containing 31 mole percent 1,4-cyclohexanedimethanol, 2) poly(1,4-cyclohexylenedi-

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methylene terephthalate) copolyesters containing 40 mole percent ethylene glycol, 3) poly(1,4-cyclohexylenedimethylene terephthalate) copolyesters containing 50 mole percent isophthalic acid, 4) 5 poly(ethylene isophthalate) copolyesters containing 50 mole percent 1,4-cyclohexanedimethanol, 5) poly(ethylene 1,4-cyclohexanedicarboxylate) copolymers containing 50 mole percent 1,4-cyclohexanedimethanol, 6) poly(ethylene terephthalate) copolymers containing 37 mole percent diethylene glycol, and the like.

The low melting, crystallizable copolyesters and substantially amorphous, high  $T_g$  copolyesters may be used in the form of powder blends or pellet blends. In general, the concentration of high  $T_g$  polyester present may range from about 1 to about 90 weight percent but the preferred range is about 5 to about 70 weight percent. A preferred blend is one wherein the crystallizable copolyester has an I.V. of 20 0.75-0.95 and is derived from 60-80 mole percent terephthalic acid, 40-20 mole percent glutaric acid, 45-65 mole percent 1,4-butanediol and 35-55 mole percent diethylene glycol, and said substantially amorphous copolyester has an I.V. of 0.45-0.80 and is 25 derived from at least 80 mole percent terephthalic acid, 60-80 mole percent ethylene glycol and 40-20 mole percent 1,4-cyclohexanedimethanol, said mole percentages being based on 100 mole percent dicarboxylic acid and 100 mole percent glycol, and each of 30 said copolyesters being present in amounts within the range of 30-70 weight percent based on their combined weight.

A typical crystallizable copolyester useful in this invention is as follows: Fifty-three and 35 thirty-five hundredths grams of dimethyl terephthalate (0.275 mole), 36 grams (0.225 mole) of

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dimethyl glutarate, 55.35 grams (0.615 mole) of 1,4-butanediol, 40.8 grams (0.385 mole) of diethylene glycol, and 1 mL of n-butanol solution of titanium tetrakisopropoxide which is 1.24 weight percent titanium are weighed into a 500-mL single-neck round-bottom flask equipped with a nitrogen inlet, stirrer, vacuum outlet, and a condensing flask. The flask is heated at 200°C for three hours with a nitrogen sweep over the reaction mixture. When the theoretical amount of methanol has distilled from the reaction mixture, the temperature is increased to 255°C at a reduced pressure of 0.5 to 0.1 mm of mercury for one hour. The flask is then removed from the heat and allowed to cool as the copolyester crystallizes. Gas chromatographic analysis of the hydrolyzed copolyester reveals the copolyester contains the following mole percentages of reaction residues: dimethyl terephthalate, 55 mole percent; dimethyl glutarate, 45 mole percent; 1,4-butanediol, 70 mole percent; and diethylene glycol, 30 mole percent. The copolyester has an inherent viscosity of 1.0 and a crystalline melting point of 115°C (DSC). The crystallization half time of this copolyester is 1.2 minutes at 60°C. The copolyester is an excellent fabric adhesive. Other crystallizable copolymers may be made using the same technique, as is well known in the art.

Typical substantially amorphous copolymers which may be used in accordance with this invention are commercially available. For example, KODAR® PETG copolyester 6763, product of Eastman Chemical Products, Inc., a copolyester of terephthalic acid and ethylene glycol modified with 1,4-cyclohexanedimethanol is particularly useful. Such substantially amorphous polyesters may be produced in accordance with known techniques, such as taught in U.S. Patent 2,901,466.

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The copolyesters prepared as described above, are individually cryogenically ground, mixed and screened using conventional procedures to obtain a powder that will pass a 70-mesh screen (200 micro-meter particles). The powders may be applied from an engraved roll to an interlining fabric to form an interlining containing rows of adhesive dots across the interlining fabric. The interlining is used to bond two different face fabrics in the examples, 10 wool/polyester and polyester/cotton. Unless otherwise indicated, the interlinings are bonded to face fabrics by pressing 10 cm squares of interlining to 10 cm squares of face fabric using either steam or an electric press. The bonded fabrics are laundered and 15 dry cleaned, then cut into strips one-inch wide and T-peel strengths reported are an average of three determinations per sample.

The crystalline melting point,  $\Delta H_f$ , and  $T_g$  referred to above are measured by conventional 20 means using a Differential Scanning Calorimeter.

Ester-forming derivatives of the acids referred to herein can be employed, if desired, to prepare the copolyesters of this invention. Examples of such ester forming derivatives are the anhydrides, 25 esters and ester chlorides of such acids.

These copolyesters are readily put into powder form using conventional grinding techniques, preferably by cryogenic grinding. The powders are nonblocking and can be readily applied to fusible 30 interlining fabrics from powder point applicators, from random sprinkling equipment, or in the form of a paste. The particles are finely divided, i.e., from about 1 micrometer to about 500 micrometers. In the application of powders from powder point applicators, 35 it is desirable to have powders with a particle size range of 50-200 micrometers (200-70 mesh). For random sprinkling application on tightly woven or non-

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woven fabrics, particle size of 150-300 micrometers (100-50 mesh) is desirable. For random sprinkling on open-weaver fabrics such as rayon/cotton blends, powders with 300-500 micrometer size (50-35 mesh) are required. For application of powder in paste form, it is necessary to have very fine powders. For example, in paste form, powder size should be 1-80 micrometer (less than 200 mesh). In the case of binder fibers, such fibers can be produced using conventional spinning techniques well known in the art. The copolyesters may also be used in other forms, such as a film.

The "heat of fusion",  $\Delta H_f$ , of polymers is the amount of heat absorbed when crystallizable polymers are melted.  $\Delta H_f$  values are readily obtained using Differential Scanning Calorimeters (DSC) (Perkins-Elmer). For example, one method for determining  $\Delta H_f$  is described in Journal of Applied Polymer Science, 20 1209 (1976). Measurement of  $\Delta H_f$  is also described in DuPont Thermal Analysis Bulletin No. 900-8 (1965). Qualitatively, it is possible to compare the degree of crystallinity of polymers by comparing their  $\Delta H_f$  values.

When copolyesters prepared as described above are employed as melt adhesives to laminate various fabric systems, metal strips and the like, excellent bonds result. These bonds are found to be highly resistant to the action of dry cleaning solvents such as perchloroethylene. The strength of the bonds is determined by a peel test based on the ASTM "T-Peel Test" set forth on pages 609-611 of the 1972 edition of the BOOK OF ASTM STANDARDS, published by the American Society for Testing Materials, and more specifically identified as Test Number D-1876-69. Results are averages of three specimens.

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If desired, dyes or dye receptive agents, color stabilizers and various other adjuvants may be added to the copolyester adhesives to meet certain specific end use requirements. Such additives would 5 normally be added as such to the polymerization mixture in which the copolyester is produced.

As used herein, the inherent viscosity (I.V.) is measured at 25°C using 0.50 gram of copolyester per 100 ml. of a solvent consisting of 60 percent by weight phenol and 40 percent by weight tetrachloroethane. 10

The following examples are submitted for a better understanding of the invention.

EXAMPLE 1

15 A poly(butylene terephthalate) copolyester containing 30 mole percent glutaric acid and 45 mole percent diethylene glycol (Polymer A) having an inherent viscosity of 0.85 dL/g (measured in 60/40 phenol-tetrachloroethane solvent) is cryogenically 20 ground in a Pulverizer with liquid nitrogen. The powder is screened through U.S. Standard sieves to give particles of 40 to 70 mesh size (coarse powder).

A poly(ethylene terephthalate) copolyester containing 30 mole percent 1,4-cyclohexanedimethanol 25 (Polymer B) having an inherent viscosity of 0.60 dL/g is cryogenically ground and sieved as described above to provide powder having a particle size of 40-70 mesh.

Equal weights (100 g) of the coarse powder 30 of each of these two copolymers are dry blended in a polyethylene bag by rotating and shaking the bag for five minutes.

The coarse powder blend is randomly sprinkled on a bonded polyester nonwoven pad to give a 35 uniform coating weight of 20 grams per 930 cm<sup>2</sup>.

The coated pad is placed in an air oven at 177°C for

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about three minutes to lightly fuse the powder. Less than 1% of the powder remains unfused after this treatment.

The coated pad 10 cm x 15 cm is bonded to a 5 10 cm x 15 cm fabric backed poly(vinyl chloride) sheeting on a Sentinil heat sealer (both platens heated; platens are 2.54 cm wide) at a bonding temperature of 155°C for 10 seconds at 1619 gm/cm<sup>2</sup> gage bonding pressure to give a 15 cm wide bond. The 10 bond is quenched immediately on a stone bench top. Five cm are trimmed from each end of the bond and three one-inch T-peel bonds are cut from each sample. These T-peel bonds are tested on an Instron machine at 20 cm/min. crosshead speed. Bonds made 15 with the powder blend have an average peel strength of 2270 gm/linear cm at 23°C and 1394 gm/linear cm at 80°C.

Similar bonds made with Polymer A coarse powder (no loss of powder after being fused on the 20 pad) give an average peel strength of 1750 gm/linear cm at 23°C but at 80°C the average peel strength is only 107 gm/linear cm.

When attempts are made to fuse unmodified coarse powder of Polymer B on the nonwoven pad, much 25 of the powder is unfused and the powder falls off the pad. This unmodified Polymer B is not useful in this bonding application.

Thus, this example illustrates that the powder blend composition provides good fusing properties and good peel strength at both 23°C and 80°C.

Equally good bonds are obtained when the nonmonomer pad is bonded to the fabric backed vinyl material using dielectric sealing equipment instead of a heat sealer.

35 EXAMPLE 2

The procedure of Example 1 is repeated except that Polymer B is replaced with a poly(1,4-

cyclohexylenedimethylene terephthalate) copolyester containing 40 mole percent ethylene glycol (Polymer C). Bonds made with a powder blend of equal weights of Polymer A and Polymer C give an average peel 5 strength of 2500 gm/linear cm at 23°C and 1770 gm/linear cm at 80°C.

EXAMPLE 3

The procedure of Example 1 is repeated except that Polymer B is replaced with a poly(ethylene 10 terephthalate) copolyester containing 37 mole percent diethylene glycol (Polymer D). Bonds made with a powder blend of equal weights of Polymer A and Polymer D give an average T-peel strength of 1966 gm/linear cm at 23°C and 1555 gm/linear cm at 80°C.

15 EXAMPLE 4

The procedure of Example 1 is repeated except that Polymer B is replaced with a poly(ethylene 20 isophthalate) copolyester containing 50 mole percent 1,4-cyclohexanedimethanol (Polymer E). Bonds made with a powder blend of equal weights of Polymers A and E give an average T-peel strength of 2502 gm/linear cm at 23°C and 2073 gm/linear cm at 80°C.

EXAMPLE 5

The procedure of Example 1 is repeated except that the powder blend is composed of 75 weight 25 percent Polymer A and 25 weight percent Polymer B. Bonds made with this blend give an average T-peel strength of 2020 gm/linear in at 23°C and 536 gm/linear cm at 80°C.

30 EXAMPLE 6

The procedure of Example 1 is repeated except that Polymer A is replaced with poly(hexamethylene terephthalate) copolyester containing 20 mole percent isophthalic acid and 20 mole percent 35 1,4-butanediol (Polymer F). Polymer F has a melting point of 104°C,  $\Delta H_f$  of 6.3 cal/g and an inherent

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viscosity of 0.72. Bonds made with a powder blend of equal weights of Polymer B and Polymer F give an average T-peel strength of 2467 gm/linear cm at 23°C and 500 gm/linear cm at 80°C.

5 EXAMPLE 7

The procedure of Example 1 is repeated except that equal weights of pellets of Polymers A and B are cryogenically coground in the pulverizer rather than being ground separately. Bonds made with coarse 10 powder (40-70 mesh) of the coground powder give an average T-peel strength of 2341 gm/linear cm at 23°C and 1376 gm/linear cm at 80°C.

EXAMPLE 8

A pellet blend (0.32 cm pellets) of 10 15 weight percent Polymer B and 90 weight percent Polymer A is prepared in a V-Cone mixer by mixing for two hours. The pellet blend is extruded into 3 mil film on a 4.5 cm, 24/1 Prodex extruder at a melt temperature of 310°F. (155°C) and with a take-up roll maintained at 80°F (27°C) The pellet blend provides film that sets up faster and gives significantly less sticking to the chill roll than that obtained with Polymer A. The faster set up of the pellet blend in extruded film form and reduced sticking to the chill 20 25 roll allows the film to be extruded at a faster rate. Polyester/cotton twill fabric is bonded with samples of this 3-mil film at 150°C on a Sentinel Heat Sealer. The bonds have a peel strength of 3664 gm/linear cm and are resistant to laundering and dry 30 cleaning treatments. Bonds were also made at 130°C and a peel strength of 2588 g/linear cm was obtained.

Similarly good results are obtained with a pellet blend of 1.0 weight percent of Polymer B and 99.0 weight percent of Polymer A.

EXAMPLE 9

The procedure of Example 1 is repeated except that the powders of Polymer A and Polymer B are sieved to a particle size of 70-200 mesh (medium powder). A powder blend of 85 weight percent of medium powder of Polymer A and 15 weight percent of medium powder of Polymer B are dry blended in a polyethylene bag by tumbling and rotating the bag for five minutes.

10 Powderpoint fusible interlinings are prepared with this medium powder blend on a small-scale laboratory powderpoint machine. The powder is applied ( $21.7 \text{ gm/m}^2$ ) from an engraved roll to a cotton interlining fabric to form an interlining containing rows of adhesive dots across the interlining fabric ( $34.9 \text{ dots/cm}^2$ ). The interlining is used to bond a polyester/cotton face fabric. The interlinings are bonded to the face fabrics by pressing four-inch squares of interlining to four-inch squares 20 of polyester twill face fabric using an electrically heated garment press at  $150^\circ\text{C}$ . The bonded fabrics are laundered and dry cleaned, then cut into strips one inch wide. T-peel strengths are determined before and after laundering and dry cleaning. The 25 T-peel strengths reported are an average of three determinations per sample. An initial peel strength of  $429 \text{ gm/linear cm}$  at  $23^\circ\text{C}$  ( $30.5 \text{ cm/minute testing rate}$ ) is observed. After commercial dry cleaning, the bonds have a peel strength of  $322 \text{ gm/linear cm}$ . 30 After laundering treatments, the bonds have a peel strength of  $322 \text{ gm/linear cm}$ . Bonds were also made at  $120^\circ\text{C}$  and a peel strength of  $268 \text{ g/linear cm}$  was obtained.

EXAMPLE 10

35 A pellet blend of 5.0 weight percent Polymer B and 95.0 weight percent Polymer A is prepared in a

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V-cone mixer by blending for two hours. The pellet blend is fed from an extruder into a gear box and through a 0.3 mm spinning orifice at 270°C. An air cavity surrounds the spinning orifice and air at the 5 temperature of the orifice is exited at high pressure at the orifice tip drawing the polymer down and cooling it and causing a melt blown web to form. The melt blown web is taken up on a silicone treated release paper. The web has a weight of 43.4 g/m<sup>2</sup>.

10 Bonds (10 cm x 15 cm) of polyester/cotton twill are made with the web at a temperature of 150°C for 15 seconds on an electricaly heated garment press. The 10 cm x 15 cm 4" x 6" bonds are cut into 2.5 cm x 15 cm pieces and tested on an Instron machine at

15 12 in/min. The results are the averages of three measurements. The web from the pellet blend gives a T-peel strength of 1609 gm/linear cm.

EXAMPLE 11

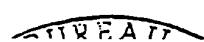
20 A polyester nonwoven web is formed by passing polyester staple fiber through a Textile Card machine to give a web weight of 13.7 gm/m<sup>2</sup>. Medium powder (70-200 mesh) of a 90/10 powder blend of Polymer A and Polymer B is fluidized with nitrogen and sprayed on the web with an electrostatic gun to give 25 a uniform coating. The web is passed through a heating chamber of infrared heaters to fuse the adhesive powder and then 3.42 through calendering rolls. The weight of adhesive in the web is 3.42 oz/yd<sup>2</sup>. The web has a nice soft hand and is strong 30 and uniformyl bonded in both the machine and transverse directions.

Unless otherwise specified, all parts, percentages, ratios, etc., are by weight.

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Claims

1. Adhesive compositions comprising a blend of
  - a) a crystallizable copolyester having a melting point of about 80-130°C and a glass transition temperature between 0°C and 20°C and
  - b) a substantially amorphous copolyester having a glass transition temperature of 50°C or higher.
- 10 2. Adhesive compositions according to claim 1 wherein said crystallizable copolyester has an I.V. of about 0.75-0.95 and is derived from about 60-80 mole percent terephthalic acid, about 40-20 mole percent glutaric acid, about 45-65 mole percent 1,4-butanediol and about 35-55 mole percent di-ethylene glycol, and said substantially amorphous copolyester has an I.V. of about 0.45-0.80 and is derived from at least 80 mole percent terephthalic acid, about 60-80 mole percent ethylene glycol and about 40-20 mole percent 1,4-cyclohexanediethanol, said mole percentages being based on 100 mole percent dicarboxylic acid and 100 mole percent glycol, and each of said copolymers being present in amounts within the range of 30-70 weight percent based on their combined weight.
- 30 3. Adhesive compositions according to claim 1 comprising about 30 to about 95 percent by weight of said crystallizable copolyester and about 5 to about 70 percent by weight of said substantially amorphous copolyester.
4. Adhesive compositions according to claim 1 in the form of a powder blend and having a particle size of about 1-500 micrometers.
- 35 5. Adhesive compositions according to claim 1 in the form of a film.

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6. Fabric material bonded together with  
the adhesive composition of claim 1.

7. Substrate material having a coating  
thereon comprising the composition of claim 1.

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# INTERNATIONAL SEARCH REPORT

International Application No PCT/US 83/01821

## I. CLASSIFICATION OF SUBJECT MATTER (If several classification symbols apply, indicate all) <sup>3</sup>

According to International Patent Classification (IPC) or to both National Classification and IPC

IPC<sup>3</sup>: C 09 J 3/16

## II. FIELDS SEARCHED

Minimum Documentation Searched <sup>4</sup>

Classification System	Classification Symbols
IPC <sup>3</sup>	C 09 J; C 08 L

Documentation Searched other than Minimum Documentation  
to the Extent that such Documents are Included in the Fields Searched <sup>4</sup>

## III. DOCUMENTS CONSIDERED TO BE RELEVANT <sup>14</sup>

Category <sup>5</sup>	Citation of Document, <sup>16</sup> with indication, where appropriate, of the relevant passages <sup>17</sup>	Relevant to Claim No. <sup>18</sup>
X	FR, A, 2073667 (DYNAMIT NOBEL) 1 October 1971, see claims 1-3 --	1-7
X	FR, A, 2426075 (RHONE POULENC) 14 December 1979, see claims 1 and 2 --	1-7
P,X	US, A, 4390668 (EDWARD B. GARVER) 28 July 1983, see claims 1-7 --	1-7
A	US, A, 4131714 (JOSEPH KAIKOSKI e.a.) 26 December 1978, see claims 1-8 cited in the application -----	1

\* Special categories of cited documents: <sup>15</sup>

"A" document defining the general state of the art which is not considered to be of particular relevance

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"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

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## IV. CERTIFICATION

Date of the Actual Completion of the International Search <sup>19</sup>

16th March 1984

Date of Mailing of this International Search Report <sup>19</sup>

18 AVR. 1984

International Searching Authority <sup>20</sup>

Signature of Authorized Officer <sup>20</sup>

INTERNATIONAL APPLICATION NO.

PCT/US 83/01821 (SA 6229)

This Annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report. The members are as contained in the European Patent Office EDP file on 05/04/84

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Patent document cited in search report	Publication date	Patent family member(s)	Publication date
FR-A- 2073667	01/10/71	NL-A- 7018062 LU-A- 62216 DE-A, B 1962642 US-A- 3699187 GB-A- 1327593 CH-A- 542913 BE-A- 760206	15/06/71 14/05/71 24/06/71 17/10/72 22/08/73 30/11/73 17/05/71
FR-A- 2426075	14/12/79	None	
US-A- 4390668	28/06/83	None	
US-A- 4131714	26/12/78	None	